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Rare-Earth Ion-Host Lattice Interactions 4. Predicting Spectra and Intensities of Lanthanides in Crystals

June 1977

U.S. Army Materiel Development and Readiness Command
HARRY DIAMOND LABORATORIES
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HDL-TR-1816—*Rare-Earth Ion-Host Lattice Interactions 4. Predicting Spectra and Intensities of Lanthanides in Crystals*,
by Clyde A. Morrison, Nick Karayianis, and Donald E. Wortman

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The spectra of Nd^{3+} and Er^{3+} in $CaWO_4$ are reanalyzed by using free-ion wave functions to determine new phenomenological crystal field parameters, B_{km}. Ratios of $[b_{km}(Er)/b_{km}(Nd)]^{1/k}$ are shown to be approximately independent of k and m. where $b_{km} = B_{km}[\langle r^k \rangle_{HF}(1 - \sigma_k)]^{-1}$. This result reinforces the previous assumption that derived B_{km} may be factored according to $B_{km} = \tau^{-k}\langle r^k \rangle_{HF}(1 - \sigma_k)A_{km}$ with the lattice sums, A_{km}, impurity

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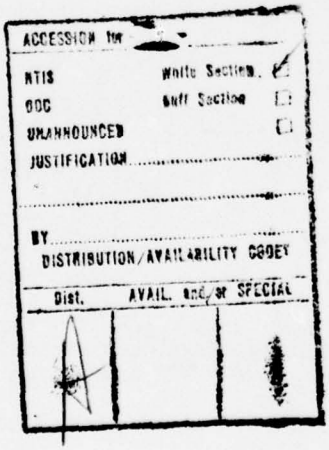
ion independent. Certain assumptions regarding excited radial wave functions in addition to these results lead to theoretical τ and $\langle r \rangle \langle r^k \rangle_{4f,n\ell}/\Delta_{n\ell}$ values for $n\ell$ = 5d, 5g for all the lanthanides. Together with $\langle r^k \rangle_{HF}$ and σ_k values obtained from the literature, one may derive B_{km} and calculate theoretical electric dipole oscillator strengths for any lanthanide-host combination where the A_{km} are determined by a lattice sum over the host's constituent ions. Tables required for performing these calculations are given.



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TABLES

1. INTRODUCTION

In the first three reports of this series,[1-3] a method was established for predicting the crystal field parameters, B_{km}, of triply ionized lanthanides in crystals. These B_{km} for even-k define the crystal field that determines the observed Stark splittings. A rationale was given for factoring the B_{km} of the lanthanide ions in a given host according to

$$B_{km} = \langle r^k \rangle (1 - \sigma_k) A_{km} , \qquad (1)$$

where the A_{km} are coefficients in the spherical tensor decomposition of the electrostatic field at the impurity ion site and are specifically impurity ion independent. Thus, it was argued, one can calculate the A_{km} by performing a lattice summation over the constituent ions of a given host material and determine the B_{km} for any lanthanide according to equation (1). If variations from host to host in the radial integrals[4] $\langle r^k \rangle$ and shielding factors[5] σ_k can be assumed small, then, presumably, B_{km} can be derived for any impurity ion-host system once $\langle r^k \rangle$ and σ_k are known for the lanthanides in some convenient host.

Phenomenological B_{km} for even-k determined by the effective spin-orbit Hamiltonian (ESOH) procedure[6] for six lanthanides in

to page 7

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[1] *N. Karayianis and C. A. Morrison, Rare Earth Ion-Host Lattice Interactions, 1. Point Charge Lattice Sum in Scheelites, Harry Diamond Laboratories TR-1648 (October 1973).*

[2] *N. Karayianis and C. A. Morrison, Rare Earth Ion-Host Crystal Interactions, 2. Local Distortion and Other Effects in Reconciling Lattice Sums and Phenomenological B_{km}, Harry Diamond Laboratories TR-1682 (January 1975).*

[3] *R. P. Leavitt, C. A. Morrison, and D. E. Wortman, Rare Earth Ion-Host Crystal Interactions, 3. Three-Parameter Theory of Crystal Fields, Harry Diamond Laboratories TR-1673 (June 1975).*

[4] *A. J. Freeman and R. E. Watson, Phys. Rev., 127 (15 September 1962), 2058.*

[5] *P. Erdös and J. H. Kang, Phys. Rev. B, 6 (1 November 1972), 3393.*

[6] *N. Karayianis, J. Chem. Phys., 53 (15 September 1970), 2460.*

$CaWO_4$ were used[3] to test this premise. It was concluded that the simplest, nontrivial forms for the $\langle r^k \rangle$ and A_{km} in equation (1) are

$$\langle r^k \rangle = \tau^{-k} \langle r^k \rangle_{HF} \tag{2}$$

and

$$A_{km} = A_{km}(q\eta) \ , \tag{3}$$

so that derived (as opposed to phenomenological) $B_{km}(\tau;q\eta)$ for even-k are functions of the three parameters, τ, q, and η. The τ parameter in this theory corrects for inadequacies in Hartree-Fock radial wave functions, and the values of τ which were found to be about 0.75 are consistent with the τ necessary to adjust Pr^{3+} Hartree-Fock values for Slater integrals to empirical values.[2] The q parameter is the effective charge on the anion in the covalent radical of the scheelite-type crystals considered (for example, the effective oxygen charge in the WO_4^{2-} radical in $CaWO_4$), and η is the ratio of the effective distance of that charge from the cation to the anion-cation nuclear distance as determined by x-ray or neutron diffraction. This so-called three-parameter theory of crystal fields was implemented[3] to determine how well one can fit derived to phenomenological B_{km} for the six lanthanides Nd, Tb, Dy, Ho, Er, and Tm in $CaWO_4$. It is important to establish how well the electrostatic field of the host material can be represented by the $A_{km}(q\eta)$, because, if the A_{km} for even-k are adequate to predict the B_{km}, then the A_{km} for odd-k--important in intensity calculations--also may be presumed adequate. It is presently impractical to obtain the latter by any other method.

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[2] *N. Karayianis and C. A. Morrison, Rare Earth Ion-Host Crystal Interactions, 2. Local Distortion and Other Effects in Reconciling Lattice Sums and Phenomenological B_{km}, Harry Diamond Laboratories TR-1682 (January 1975).*

[3] *R. P. Leavitt, C. A. Morrison, and D. E. Wortman, Rare Earth Ion-Host Crystal Interactions, 3. Three-Parameter Theory of Crystal Fields, Harry Diamond Laboratories TR-1673 (June 1975).*

Point charge lattice sums $A_{km}(q\eta)$ were performed,[3] and τ, q, and η were varied to fit derived to phenomenological B_{km} independently for the six ions in $CaWO_4$. It was then shown, by fixing the effective oxygen charge to q = -1.09 and its fractional distance from tungsten to η = 0.977, that a best τ for each of the six lanthanides could be determined that gives substantially the same fit to phenomenological B_{km} as previously obtained when τ, q, and η were varied independently. The fact that fits to the B_{km} of each ion could be made with the same A_{km} (-1.09, 0.977) represents a qualified success for the theory, but the rms deviations remaining between derived and phenomenological B_{km} are still too large, ranging from 13 (for Er) to 135 cm^{-1} (for Tb), to give one confidence in the predicting capabilities of the theory.

cont from p. 5

In this report, the latest values for quantities necessary to derive the B_{km} are used in an attempt to isolate the source of these large discrepancies. A new analysis using free-ion wave functions is performed on the spectra of Nd and Er in $CaWO_4$ to determine more accurate tau factors for these ions. A linear interpolation to all the lanthanides of these τ, along with smoothed values for Freeman and Watson's[4] $\langle r^k \rangle_{HF}$ and linearly interpolated values for the σ_k of Erdös and Kang,[5] are used to determine ρ_k, where

$$\rho_k(\tau) = \tau^{-k} \langle r^k \rangle_{HF} (1 - \sigma_k) , \tag{4}$$

so that

$$B_{km}(\tau;q\eta) = \rho_k(\tau) A_{km}(q\eta) . \tag{5}$$

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[3] *R. P. Leavitt, C. A. Morrison, and D. E. Wortman, Rare Earth Ion-Host Crystal Interactions, 3. Three-Parameter Theory of Crystal Fields, Harry Diamond Laboratories TR-1673 (June 1975).*

[4] *A. J. Freeman and R. E. Watson, Phys. Rev., 127 (15 September 1962), 2058.*

[5] *P. Erdös and J. H. Kang, Phys. Rev. B, 6 (1 November 1972), 3393.*

Values are derived also for quantities necessary for calculating transition probabilities such as $\langle r^p \rangle_{4f,n\ell}$ for p = 1, 3, 5, and 7 and $\Delta_{n\ell} = E_{n\ell} - E_{4f}$ for $n\ell$ = 5d and 5g.

The results given in this report set the stage for calculating spectra and electric dipole transition probabilities effectively from first principles for all the lanthanides. These calculations will be performed for many of the current experimental host materials to establish an initial competency in predicting potential laser properties of new materials. Results of this project will be the topic of subsequent reports in this series. While it may be overly optimistic to expect that one can accurately predict spectra once the crystal coordinates of the host material are known, it is hoped that some guidance can be given as to which materials show greatest promise as laser materials.

2. DERIVING CRYSTAL FIELD PARAMETERS

The first step in deriving the ρ_k for the lanthanides according to equation (4) requires a determination of $\langle r^k \rangle_{HF}$ and σ_k. We have obtained fifth-degree polynomial expressions in N that fit the values of $\langle r^k \rangle_{HF}$ calculated by Freeman and Watson[4] for Ce, Pr, Nd, Sm, Dy, Er, and Yb to better than 0.24 percent accuracy. Letting

$$\langle r^k \rangle_{HF} = \sum_{m=0}^{5} r_{km} N^m , \tag{6}$$

where N is the number of f electrons for a given triply ionized lanthanide, one gets values for the r_{km} given in table I. These

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[4] *A. J. Freeman and R. E. Watson, Phys. Rev., 127 (15 September 1962), 2058.*

TABLE I. VALUES FOR r_{km} IN UNITS OF $Å^k$ WHERE $\langle r^k \rangle_{HF} = \Sigma_m r_{km} N^m$

m	r_{2m}	r_{4m}	r_{6m}
0	3.79955(-1)	3.45431(-1)	6.56496(-1)
1	-5.13951(-2)	-9.04155(-2)	-2.36090(-1)
2	8.32091(-3)	1.78303(-2)	5.12945(-2)
3	-8.92996(-4)	-2.05603(-3)	-6.15291(-3)
4	5.02302(-5)	1.20584(-4)	3.69091(-4)
5	-1.12879(-6)	-2.78753(-6)	-8.64990(-6)

NOTE: Numbers in parentheses are powers of 10.

formulas give values for Gd(N = 7) of $\langle r^k \rangle_{HF}$ = 0.223, 0.124, and 0.148 in units of $Å^k$ for k = 2, 4, and 6, respectively, whereas Freeman and Watson obtain 0.220, 0.119, and 0.138 by their Hartree-Fock method.* The differences between corresponding values indicate the degree of accuracy that one may expect in extending the polynomial fits to the lanthanides for which radial integrals were not obtained by Freeman and Watson. For completeness, table II gives the $\langle r^k \rangle_{HF}$ for all the triply ionized lanthanides calculated from equation (6) by using the r_{km} values in table I.

TABLE II. VALUES FOR τ, $\langle r^k \rangle_{HF}$ (Åk) AND σ_k IN $\rho_k = \tau^{-k} \langle r^k \rangle_{HF} (1 - \sigma_k)$ WHERE DERIVED CRYSTAL FIELD PARAMETERS ARE GIVEN BY $B_{km} = \rho_k A_{km}$ FOR GROUND f^N CONFIGURATIONS OF TRIPLY IONIZED LANTHANIDES

Ion	N	τ	$\langle r^2 \rangle_{HF}$	$\langle r^4 \rangle_{HF}$	$\langle r^6 \rangle_{HF}$	σ_2	σ_4	σ_6	ρ_2	ρ_4	ρ_6
Ce	1	0.7693	0.3360	0.2709	0.4659	0.6757	0.0254	-0.0422	0.1841	0.7536	2.3417
Pr	2	0.7597	0.3041	0.2213	0.3459	0.6667	0.0272	-0.0421	0.1756	0.6464	1.8754
Nd	3	0.7500	0.2803	0.1882	0.2715	0.6578	0.0290	-0.0420	0.1706	0.5776	1.5897
Pm	4	0.7403	0.2621	0.1655	0.2247	0.6488	0.0308	-0.0418	0.1679	0.5339	1.4213
Sm	5	0.7306	0.2472	0.1488	0.1929	0.6398	0.0327	-0.0417	0.1668	0.5049	1.3210
Eu	6	0.7210	0.2347	0.1353	0.1686	0.6309	0.0345	-0.0415	0.1666	0.4836	1.2503
Gd	7	0.7113	0.2232	0.1237	0.1477	0.6220	0.0363	-0.0414	0.1668	0.4656	1.1873
Tb	8	0.7016	0.2129	0.1131	0.1287	0.6130	0.0381	-0.0413	0.1673	0.4990	1.1232
Dy	9	0.6919	0.2033	0.1037	0.1119	0.6041	0.0399	-0.0411	0.1681	0.4341	1.0614
Ho	10	0.6823	0.1945	0.0954	0.0981	0.5951	0.0418	-0.0410	0.1692	0.4217	1.0119
Er	11	0.6726	0.1865	0.0883	0.0874	0.5861	0.0436	-0.0408	0.1706	0.4126	0.9826
Tm	12	0.6629	0.1790	0.0820	0.0787	0.5772	0.0454	-0.0407	0.1722	0.4053	0.9649
Yb	13	0.6532	0.1717	0.0753	0.0681	0.5683	0.0472	-0.0406	0.1737	0.3938	0.9120

Values for the σ_k were obtained by linear interpolation by using the Erdös and Kang[5] values for Pr and Tm. One gets

**Multiply the values of Freeman and Watson, which are in atomic units, by $(0.529172)^k$ to obtain $Å^k$ units, where the angstrom unit $Å = 10^{-8}$ cm.*

[5] *P. Erdös and J. H. Kang, Phys. Rev. B, 6 (1 November 1972), 3393.*

$$\sigma_2 = 0.6846 - 0.00895N \ , \tag{7a}$$

$$\sigma_4 = 0.02356 + 0.00182N \ , \tag{7b}$$

$$\sigma_6 = -0.04238 + 0.00014N \ , \tag{7c}$$

which generate the values given in table II.

Values for the τ given in table II are calculated by the formula

$$\tau = 0.75(1.0387 - 0.0129N) \ , \tag{8}$$

which was obtained by a linear interpolation of values obtained for Nd^{3+} and Er^{3+} in $CaWO_4$ as follows: Free-ion basis wave functions[7] for the lowest 14 and 10 multiplets, respectively, for Nd and Er were obtained by using the "free-ion" parameters of Carnall, Fields, and Rajnak.[7] Crystal field subspaces for S_4 symmetry were then set up and diagonalized for the crystal field Hamiltonian

$$H_x = \Sigma_{km} B_{km}^{\dagger} \Sigma_i C_{km}(\hat{r}_i) \ ,$$

where the C_{km} are unnormalized spherical harmonics. By varying the B_{km} and the multiplet centroids, fits of 4.56 and 3.80 cm^{-1} rms were obtained,[8] respectively, to 51 (out of 64) and 38 (out of 58) experimentally identified Stark levels of Nd and Er. These parameters are given in table III, columns 2 and 4. If the phenomenological B_{km} are factorable according to equations (4) and (5) where the A_{km} are impurity ion independent, then b_{km}, defined by

$$b_{km} = B_{km}\left[\langle r^k \rangle_{HF} \left(1 - \sigma_k\right)\right]^{-1} , \tag{9}$$

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[7] *W. T. Carnall, P. R. Fields, and K. Rajnak, J. Chem. Phys., 49 (1968), 4424.*

[8] *D. E. Wortman, C. A. Morrison and N. Karayianis, Rare Earth Ion-Host Lattice Interactions, 5. Lanthanides in $CaWO_4$, Harry Diamond Laboratories TR-1794 (June 1977).*

by using the $\langle r^k \rangle_{HF}$ and σ_k values from table II, and evaluated for two different ions, will be related according to

$$\left[b_{km}(Er)/b_{km}(Nd)\right]^{1/k} = \tau_{Nd}/\tau_{Er} \ , \tag{10}$$

independently of k and m. The b_{km} and τ_{Nd}/τ_{Er} values given in table III demonstrate the approximate constancy in the latter values with the possible exception of the value calculated from b_{20}. Giving double weight to the 4,4 and 6,4 components, we obtain approximately

$$\tau_{Er} = \tau_{Nd}/1.115 \tag{11}$$

from the average of the values in table III, column 6.

TABLE III. PHENOMENOLOGICAL B_{km} FOR Nd^{3+} AND Er^{3+} IN $CaWO_4$ AND CALCULATED $b_{km} = B_{km}\left[\langle r^k \rangle_{HF} (1 - \sigma_k)\right]^{-1}$

	Nd		Er			Nd (derived)	
km	B_{km}	b_{km}	B_{km}	b_{km}	$\left[b_{km}(Er)/b_{km}(nd)\right]^{1/k} = \tau_{Nd}/\tau_{Er}$	$B_{km}(0.752, -1.15, 0.962)$	ΔB_{km}
20	503	5244	433	5610	1.0343	393	110
40	-878	-4806	-655	-7756	1.1271	-1062	184
44	1042	5703	809	9580	1.1385	1119	77
60	-3.84	-13.57	-2.56	-28.14	1.1293	5.58	9
464	905	3199	546	6002	1.1106	734	171
164	238	841	156	1715	1.1261	41	197
					$\overline{1.115}$ weighted av		$\overline{141}$ rms

NOTE: Appropriate values are used from table II. Derived $B_{km}(t;q,n)$ and absolute differences between derived and phenomenological values for Nd are given in the last two columns.

To obtain values for each τ, we performed a lattice sum over the $CaWO_4$ ion setting, in units of proton charge, $q_{Ca} = +2$ and $q_W = -2 - 4q$ and varied the oxygen charge, q, its effective fractional distance from

tungsten, η, and τ_{Nd} to obtain a best fit to the phenomenological B_{km} of Nd. The resulting values for τ, q, and η, which are

$$\tau_{Nd} = 0.75 , \tag{12a}$$

$$q = -1.150 , \tag{12b}$$

$$\eta = 0.962 , \tag{12c}$$

gave a fit of 141 cm^{-1} rms between derived (table III, column 7) and phenomenological (table III, column 2) B_{km} for Nd^{3+} in $CaWO_4$. This "fit" is worse than that obtained to the phenomenological B_{km} resulting from the inherently less-accurate ESOH method.[3] Assuming a linear variation for τ over the lanthanides, equations (12a) and (11) give equation (8) and the calculated values for τ in table II, column 3. According to equation (4), one may then calculate the values for ρ_k given in the last three columns of table II.

The main ingredients for deriving B_{km} for any lanthanide-host combination have thus been obtained, provided that one assumes that the τ, $\langle r^k \rangle_{HF}$, and σ_k are approximately host independent. The final task in deriving B_{km} in a given host, under these assumptions, is to develop an adequate model of the particular host material for calculating the A_{km}--the q,η model assumed in the three-parameter theory is but an initial effort in this direction. The large discrepancy of 141 cm^{-1} rms between derived and phenomenological B_{km} for Nd^{3+} in $CaWO_4$ suggests that further theoretical work is required to determine the ultimate validity of this theoretical approach.

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[3] *R. P. Leavitt, C. A. Morrison, and D. E. Wortman, Rare Earth Ion-Host Crystal Interactions, 3. Three-Parameter Theory of Crystal Fields, Harry Diamond Laboratories TR-1673 (June 1975).*

3. CALCULATING ELECTRIC DIPOLE INTENSITIES

To calculate electric dipole intensities,[9] one must know A_{km} values for odd-k, r^k expectation values between the ground 4f and excited $n\ell$ configurations, where $\ell + 3 =$ odd, and energy differences between the configurations $\Delta_{n\ell} = E_{n\ell} - E_{4f}$. Specifically, the matrix element $M_\mu(ij)$ that determines the electric dipole intensity of polarization μ between the states i and j in the ground f^N configuration is given by

$$M_\mu(ij) = \sum_{Jkmn\ell} A^+_{km} \langle r \rangle_{4f,n\ell} \left\langle r^k \right\rangle_{4f,n\ell} \Delta^{-1}_{n\ell} \left\langle i \left| U^{(J)}_{m+\mu} \right| j \right\rangle$$

$$\times \left\{ [3][\ell][J] \right\}^{\frac{1}{2}} \left\langle 3(0)1(0) \middle| \ell(0) \right\rangle \left\langle \ell(0)k(0) \middle| 3(0) \right\rangle \tag{13}$$

$$\left\{ W(3k3l;\ell J) \left\langle k(m)1(\mu) \middle| J(m+\mu) \right\rangle \right. ,$$

where

$U^{(J)}$ is a unit spherical tensor for N equivalent electrons,[10]

$[x] \equiv 2x + 1$,

$\left\langle j_1(m_1) j_2(m_2) \middle| j(m) \right\rangle$ is a Clebsch-Gordan coefficient,[11]

and W(abcd;ef) is a Racah coefficient.[11]

The oscillator strength P_μ of the transition is given by[9,12]

$$P_\mu = \chi \left[4\pi m_e \nu_{ij} / \hbar \right] M^2_\mu(ij) \ , \tag{14}$$

---

[9] *B. R. Judd, Phys. Rev., 127 (1 August 1962), 750. Note that our A^+_{km} are Judd's A_{km}.*

[10] *C. W. Nielson and G. F. Koster, Spectroscopic Coefficients for the p^n, d^n and f^n Configurations, The MIT Press, Cambridge, MA (1963).*

[11] *M. Rotenberg, R. Bivins, N. Metropolis, and J. K. Wooten, The 3-j and 6-j Symbols, The Technology Press, Cambridge, MA (1959). See their p. 1, eq (1.1) to relate Clebsch-Gordan coefficients to 3-j symbols and p. 13, eq. (2.1) to relate Racah coefficients to 6-j symbols.*

[12] *M. J. Weber, T. E. Varitimos and B. H. Matsinger, Phys. Rev. B, 8 (1 July 1973), 47.*

where, in terms of the refractive index n, $\chi = n(n^2 + 2)^2/9$, m_e is the electron mass, ν_{ij} is the frequency of the radiation, and $\hbar$ is Planck's constant divided by 2π.

As is customary, we assume that mixing between the 4f and the excited 5d and 5g configurations is primarily responsible for electric dipole transitions. To obtain approximate values for the radial integral between these configurations, we assume orthogonalized hydrogen-like radial wave functions. The wave functions are defined by comparing expectation values with experimental and theoretical data from the literature wherever possible.

We choose 4f, 5d, and 5g normalized wave functions as follows:

$$\psi_{4f} = W_{4f} r^4 \exp(-yr) \ , \tag{15a}$$

$$\psi_{5d} = W_{5d}(r^5 + Ar^4 + Br^3)\exp(-xr) \ , \tag{15b}$$

$$\psi_{5g} = W_{5g}(r^5 + Cr^4)\exp(-xr) \ , \tag{15c}$$

where

$$\int_0^\infty dr \ \psi_i \psi_j = \delta_{ij} \ . \tag{16}$$

From equation (16), the normalizations and the coefficients A, B, and C are defined in terms of y and x (see app A).

To determine y as a function of N, the number of electrons in the ground f^N configuration of a triply ionized lanthanide, ratios of $\langle r^5 \rangle_{4f,4f} / \langle r^3 \rangle_{4f,4f,} = 39/y^2$ were adjusted to fit similar ratios calculated by Freeman and Watson using their Hartree-Fock wave functions.[4] The quadratic relationship

$$y(N) = 5.53358 + 0.228834N - 0.00482594N^2 \ \text{Å}^{-1} \tag{17}$$

---

[4] *A. J. Freeman and R. E. Watson, Phys. Rev., 127 (15 September 1962), 2058.*

gives fits of 1.03 percent or better to the eight calculated ratios of Freeman and Watson, as shown in table IV.

TABLE IV. RATIO OF FREEMAN AND WATSON HARTREE-FOCK EXPECTATION VALUES FOR SEVERAL TRIPLY IONIZED LANTHANIDE GROUND f^N CONFIGURATIONS

N	$\langle r^5\rangle_{4f,4f}/\langle r^3\rangle_{4f,4f}$ ($Å^2$)	γ_{HF} ($Å^{-1}$)	$\gamma(N)$ ($Å^{-1}$)	$39[\gamma(N)]^{-2}$ ($Å^2$)
1	1.20136	5.69850	5.75759	1.17648
2	1.08628	5.99263	5.97194	1.09354
3	1.00510	6.22944	6.17665	1.02225
5	0.90346	6.57004	6.55710	0.90707
7	0.81256	6.92778	6.89895	0.81941
9	0.76492	7.14052	7.20219	0.75186
11	0.70666	7.42873	7.46682	0.69951
13	0.65160	7.73645	7.69284	0.65901

NOTE: Effective hydrogenic wave function exponent in $\psi_{4f} \sim \exp(-\gamma_{HF} r)$, quadratic best fit $\gamma(N) = 5.53358 + 0.228834N - 0.00482594N^2$, and corresponding ratios assuming $\psi_{4f} \sim \exp[-\gamma(N)r]$.

To determine x, calculated ratios of $\langle r^5\rangle_{4f,5d}/\langle r^3\rangle_{4f,5d}$ were fit to similar ratios calculated from Grossgut's[13] wave functions for Ce^{3+}, Pr^{3+}, and Nd^{3+}. A linear variation of x according to

$$x(N) = 2.48605 + 0.044175N \text{ Å}^{-1} \tag{18}$$

gives a fit to Grossgut's ratios of 2.00 percent or better as shown in table V.

The complete set of calculated y, x, and expectation values for all the triply ionized lanthanides is given in table VI where columns b, c, d, and e are normalized to column a for each N.

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[13] *P. Grossgut, Analytical Wave Functions and Oscillator Strengths for Ce^{3+}, Pr^{3+}, Nd^{3+} and Cu^{2+}, Ph.D Dissertation, Texas Christian University (1971) (University Microfilms, Ann Arbor MI, No. 72-7621).*

TABLE V. RATIO OF GROSSGUT'S HARTREE-FOCK EXPECTATION VALUES FOR Ce, Pr, AND Nd GROUND f^N CONFIGURATIONS

N	$\langle r^5\rangle_{4f,5d}/\langle r^3\rangle_{4f,5d}$ ($Å^2$)	x_{HF} ($Å^{-1}$)	x(N) ($Å^{-1}$)	$r^5\rangle/\langle r^3\rangle_{x(N)}$ ($Å^2$)
1	2.20785	2.55597	2.53023	2.23898
2	2.19145	2.52291	2.57440	2.13541
3	2.01422	2.64432	2.61858	2.03955

NOTE: "Effective" hydrogenic wave-function exponent in $\psi_{5d} \sim \exp(-x_{HF}r)$ assuming $\psi_{4f} \sim \exp(-y(N)r)$ with y(N) from table IV, linear best fit x(N) = 2.48605 + 0.044175 N, and ratio of expectation values calculated with x(N) for comparison with Grossgut's values.

TABLE VI. BEST-FIT EXPONENTIAL FACTORS IN $\psi_{4f} \sim \exp[-y(N)r]$ AND ψ_{5d}

N	y(N) ($Å^{-1}$)	x(N) ($Å^{-1}$)	a ($Å^{-1}$)	b[b] ($Å^{-1}$)	c[c] (1)	d[d] ($Å^{-1}$)	e[e] ($Å^{-1}$)
1	5.75759	2.53023	0.11491	2.23898	0.202621	0.766971	3.28281
2	5.97194	2.5744	9.40786E-2	2.13541	0.188527	0.671099	2.7013
3	6.17665	2.61858	8.03255E-2	2.03955	0.177361	0.596127	2.26565
4	6.3717	2.66275	6.93901E-2	1.9512	0.168458	0.536615	1.93289
5	6.5571	2.70693	6.06361E-2	1.86996	0.161347	0.488805	1.67449
6	6.73285	2.7511	5.35826E-2	1.79534	0.155688	0.450039	1.47102
7	6.89895	2.79528	4.78655E-2	1.72685	0.151232	0.418399	1.30892
8	7.05539	2.83945	4.32078E-2	1.66396	0.147794	0.392475	1.17853
9	7.20218	2.88363	3.93978E-2	1.60621	0.145236	0.371216	1.07288
10	7.33933	2.9278	3.62731E-2	1.55314	0.143456	0.35383	0.986835
11	7.46682	2.97197	3.37077E-2	1.50434	0.142378	0.339716	0.916565
12	7.58465	3.01615	3.16041E-2	1.45944	0.141947	0.328415	0.859198
13	7.69284	3.06032	0.029886	1.41808	0.142127	0.319578	0.812554

NOTE:
[a] $\psi_{5d} \sim \exp[-x(N)r]$ from tables IV and V and calculated expectation values and ratios from these wave functions.
[b] Multiply by corresponding values in column "a" to obtain $\langle r\rangle\langle r^2\rangle$ values:
a: $\langle r\rangle_{4f,5d}\langle r^3\rangle_{4f,5d}$
b: $\langle r\rangle_{4f,5d}\langle r^3\rangle_{4f,5d}$/(values in col. a)
c: $\langle r\rangle_{4f,5d}\langle r^3\rangle_{4f,5d}$/(values in col. a)
d: $\langle r\rangle_{4f,5d}\langle r^3\rangle_{4f,5d}$/(values in col. a)
e: $\langle r\rangle_{4f,5d}\langle r^3\rangle_{4f,5d}$/(values in col. a)

The final factors required for electric dipole intensity calculations are the energy differences $\Delta_{n\ell} = E_{n\ell} - E_{4f}$ for $n\ell$ = 5d and

5g.[14-16] Table VII is self-explanatory as to the origin of the values we have chosen. With these $\Delta_{n\ell}$ and the $\langle r \rangle \langle r^k \rangle$ of table VI, we obtain the final normalized factors of $\langle r \rangle \langle r^k \rangle / \Delta_{n\ell}$ given in table VIII. These values should be multiplied by the corresponding factor in table VI, column a, for each N if intensity comparisons between ions are to be made.

TABLE VII. ENERGIES IN 10^3 cm^{-1} FOR 4f, 5d, AND 5g, DIFFERENCES $\Delta_{5d} = E_{5d} - E_{4f}$ (AND SIMILARLY FOR 5g), AND RATIOS FOR THE TRIPLY-IONIZED LANTHANIDES

Ion	$(-E_{4f})^a$	$(-E_{5d})^b$	$(-E_{5g})^c$	$(\Delta_{5d})^d$	$(\Delta_{5g})^e$	Δ_{5g}/Δ_{5d}
Ce	296.5	246.8	74.0	49.7*	222.5	4.48
Pr	314.4	253.2	76.0	61.2*	238.4	3.90
Nd	325.2	254.8	76.4	70.4	248.8	3.53
Pm	328.2	256.6	77.0	71.6	251.2	3.51
Sm	330.8	258.3	77.5	72.5	253.3	3.49
Eu	341.0	260.0	78.0	81.0	263.0	3.25
Gd	353.8	261.5	78.4	92.3*	275.4	2.98
Tb	318.6	263.5	79.0	55.1	239.6	4.35
Dy	331.9	265.3	79.6	66.6	252.3	3.79
Ho	341.6	267.0	80.1	74.6	261.5	3.51
Er	342.6	268.7	80.6	73.9	262.0	3.55
Tm	343.2	270.5	81.2	72.7	262.0	3.60
Yb	352.1	272.2	81.7	79.9	270.4	3.38

*Measured values. See footnote a below.

[a] K. L. Vander Sluis and L. J. Nugent, J. Chem. Phys., 60 (1974), 1928, table II.

[b] Calculated by subtracting col. d from col. a so that $-E_{5d} = -E_{5f} - \Delta_{5d}$.

[c] Calculated by $-E_{5g} = 0.3(-E_{5d})$, which gives $E_{5g}(Nd) \approx -76{,}400\ cm^{-1}$--a value similar to that obtained from the Lu III spectrum from V. Kaufman and J. Sugar, J. Opt. Soc. Am., 61 (1971), 1693, as follows: From $E_{6s} = (E_{6s} - E_{5g}) - 9R/(n^*_{5g})^2$, $E_{6s} = -169.049$ and $E_{6s} - E_{5g} = -129.107$ in units of $10^3\ cm^{-1}$, then $n^*_{5g} = 4.9726$. Thus, $T_{5g} = 16R/(n^*_{5g})^2 = 71{,}000\ cm^{-1} \sim -E_{5g}$ for all the lanthanides.

[d] K. L. Vander Sluis and L. J. Nugent, J. Chem. Phys., 60 (1974), 1927, table I.

[e] Calculated by subtracting col. c from col. a so that $\Delta_{5g} = -E_{4f} + E_{5g}$.

[14] K. L. Vander Sluis and L. J. Nugent, J. Chem. Phys., 60 (1 March 1974), 1927; J. Opt. Soc. Am., 64 (May 1974), 687.

[15] V. Kaufman and J. Sugar, J. Opt. Soc. Am., 61 (1971), 1693.

[16] J. Sugar and J. Reader, J. Chem. Phys., 59 (15 August 1973), 2083.

TABLE VIII. NORMALIZED $\langle r \rangle \langle r^k \rangle / \Delta_{5\ell}$ VALUES FOR TRIPLY-IONIZED LANTHANIDES

N	Ion	a*	b*	c*	d*	e*
1	Ce	2.01207	4.50499	0.0910822	0.344768	1.475690
2	Pr	1.63399	3.48924	0.0790669	0.281454	1.132910
3	Nd	1.42045	2.89709	0.0712982	0.239639	0.910776
4	Pm	1.39665	2.72514	0.0670561	0.213604	0.769400
5	Sm	1.37931	2.57925	0.0636956	0.192967	0.661045
6	Eu	1.23457	2.21647	0.0591970	0.171118	0.559324
7	Gd	1.08342	1.87091	0.0549235	0.151952	0.475365
8	Tb	1.81488	3.01990	0.0616965	0.163839	0.491978
9	Dy	1.50150	2.41173	0.0575626	0.147127	0.425224
10	Ho	1.34048	2.08196	0.0548590	0.135308	0.377375
11	Er	1.35318	2.03565	0.0543490	0.129668	0.349847
12	Tm	1.37552	2.00748	0.0541681	0.125325	0.327876
13	Yb	1.25156	1.77482	0.0525539	0.118170	0.300456

NOTE:
Normalized by using $\Delta_{5\ell}$ (ℓ = d, g) from table VII and $\langle r \rangle \langle r^k \rangle$ from table VI.
**Multiply by corresponding values in table IV, col. a, and divide by 100 to obtain actual values.*
[a] *Normalized* $\langle r \rangle \langle r^3 \rangle / \Delta_{5d}$
[b] *Normalized* $\langle r \rangle \langle r^5 \rangle / \Delta_{5d}$
[c] *Normalized* $\langle r \rangle \langle r^3 \rangle / \Delta_{5g}$
[d] *Normalized* $\langle r \rangle \langle r^5 \rangle / \Delta_{5g}$
[e] *Normalized* $\langle r \rangle \langle r^7 \rangle / \Delta_{5g}$

4. DISCUSSION AND CONCLUSIONS

It is important to note that the ostensibly superior method described in this report of using free-ion wave functions to determine phenomenological B_{km} for Nd^{3+} in $CaWO_4$ resulted in a fit (141 cm^{-1} compared with 101 cm^{-1} rms) between phenomenological and derived B_{km} inferior to that obtained previously[3] (by using Russell-Saunders wave functions in the ESOH method). This result does not indicate that the discrepancy in the fits is necessarily due to

[3] *R. P. Leavitt, C. A. Morrison, and D. E. Wortman, Rare Earth Ion-Host Crystal Interactions, 3. Three-Parameter Theory of Crystal Fields, Harry Diamond Laboratories TR-1673 (June 1975).*

incorrect phenomenological B_{km}, but that further theoretical work probably is required to determine more accurate values for derived B_{km}.

On the other hand, a comparison of the Nd results with phenomenological B_{km} determined for Er^{3+} in $CaWO_4$, also using free-ion wave functions, resulted in an approximate constancy with respect to k and m in values for $\left[b_{km}(Er)/b_{km}(Nd)\right]^{1/k}$. Thus, the crucial assumption in the theory remains so far intact--that the A_{km} are impurity ion independent. Without this assumption, it would be very difficult to continue the search for a theoretical model to predict impurity ion spectra.

There are several areas for improvements in the theory that readily come to mind. First, better values for $\langle r^k \rangle$ and σ_k may be determined by including electron correlations[17] and relativistic corrections[18] in the radial wave functions--effects neglected in the Hartree-Fock approximation. Second, one may determine the homogeneous dielectric effect of placing the impurity ion in a host environment and also the wave-function overlap effects (which, incidently, might in part be contained implicitly in the dielectric calculation). Third, and probably most important, the dipolar (and possibly higher multipolar) contributions to the A_{km} in the host material should be considered, since their contribution has already been shown to be substantial.[19]

The ρ_k (table II) and $\langle r \rangle \langle r^k \rangle / \Delta$ values (table VIII), while probably not sufficient for predicting accurate spectra and electric dipole

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[17] *J. C. Morrison, J. Phys. B: Atom. Molec. Phys., 6 (November 1973), 2205.*

[18] *W. B. Lewis, Relativistic Calculations of $\langle r^{-3} \rangle$ and Other $\langle r^n \rangle$ Parameters Encountered in Magnetic Resonance of Rare-Earth Ions and Atoms, Proc. XVIth Congress A.M.P.E.R.E., Bucharest, Rumania (1 to 5 September 1970), (Publishing House of Academy of Socialist Republic of Rumania, 1971).*

[19] *C. A. Morrison, Solid State Commun., 18 (1976), 153.*

intensities, nevertheless will be used exclusively in the following 10 reports in this series to interpret and predict spectra for the lanthanides in $CaWO_4$, $LiYF_4$, YVO_4, YPO_4, $YAsO_4$, $Y_2SiBe_2O_7$, $Y_3Al_5O_{12}$ (YAG), $Y_3Ga_5O_{12}$ (YGaG), $YAlO_3$, and LnP_5O_{14}, where Ln is any of the lanthanides. These calculations will enable one to identify the specific areas of deficiency in the present theory by comparing the predictions with the mass of reported data in the literature.

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APPENDIX A.--HYDROGEN-LIKE WAVE FUNCTIONS AND EXPECTATION VALUES

From equation (16) in the main body of the report for i = j, one obtains

$$W_{4f} = \frac{2y^4}{3}\left(\frac{y}{35}\right)^{\frac{1}{2}}, \tag{A-1}$$

$$W_{5d} = \frac{2x^5}{45}\left(\frac{2x}{7}\right)^{\frac{1}{2}}\left[1 + A\,\frac{2x}{5} + (A^2 + 2B)\,\frac{2x^2}{45} + AB\,\frac{x^2}{45} + B^2\,\frac{x^4}{315}\right]^{-\frac{1}{2}}, \tag{A-2}$$

$$W_{5g} = \frac{2x^5}{45}\left(\frac{2x}{7}\right)^{\frac{1}{2}}\left[1 + C\,\frac{2x}{5} + C^2\,\frac{2x^2}{45}\right]^{-\frac{1}{2}}. \tag{A-3}$$

From the same equation for $i \neq j$, one has

$$C = -\frac{9}{y + x}, \tag{A-4}$$

$$A = \left[\frac{8}{5}\left(\frac{x}{y + x}\right)^2\left(2 + C\,\frac{x}{2}\right) - 1 - C\,\frac{x}{5}\right] \times \left[\left(1 + C\,\frac{2x}{9}\right)\frac{x}{5} + C\,\frac{8}{5}\left(\frac{x}{y + x}\right)^2\left(2 + C\,\frac{x}{2}\right)\right]^{-1}, \tag{A-5}$$

$$B = -C^2\,\frac{8}{9}\left(1 - \frac{a}{C}\right), \tag{A-6}$$

all in terms of the exponential factors y and x, respectively, for the 4f and both the 5d and 5g wave functions. The required r^p expectation values in these states are given by

$$\langle r^p \rangle_{4f,4f} = W_{4f}^2\,\frac{(8 + p)!}{(2y)^{9+p}}, \tag{A-7}$$

$$\langle r^p \rangle_{4f,5d} = W_{4f}W_{5d}\left[\frac{(9 + p)!}{(y + x)^{10+p}} + A\,\frac{(8 + p)!}{(y + x)^{9+p}} + B\,\frac{(7 + p)!}{(y + x)^{8+p}}\right], \tag{A-8}$$

$$\left\langle r^p \right\rangle_{4f,5g} = W_{4f} W_{5g} \left[\frac{(9 + p)!}{(y + x)^{10+p}} + C \frac{(8 + p)!}{(y + x)^{9+p}} \right] . \tag{A-9}$$

Thus, all of the normalizations of the radial wave functions, as well as the coefficients A, B, and C defining them (according to eq (15a) to (15c)), are given once y and x are determined. From table VI in the main body of the report, one has for Nd, for example,

$$\psi_{4f}(\mathrm{Nd}) \sim r^4 \exp(-6r) , \tag{A-10}$$

$$\psi_{5d}(\mathrm{Nd}) \sim (r^5 - 3r^4 + 2r^3)\exp(-3r) , \tag{A-11}$$

$$\psi_{5g}(\mathrm{Nd}) \sim (r^5 - r^4)\exp(-3r) . \tag{A-12}$$

In retrospect, it might have been better to choose the ψ_{5g} wave function as pure hydrogenic as $\psi_{5g} \sim r^5 \exp(-xr)$ and to throw the deviation from hydrogen-like behavior in the ψ_{4f} wave function, because the former--being farther removed from the core--is expected to be more hydrogen-like. However, since the wave functions are used to calculate only ratios of expectation values that are important in determining relative electric dipole intensities, the ratios are not expected to be sensitive to this choice.

A further question that may legitimately be asked is why all three radial wave functions were chosen mutually orthogonal. It is true that hydrogenic radial wave functions are orthogonal either within an "n" shell $\left\langle \psi_{n\ell} \psi_{n\ell'} \right\rangle = \delta_{\ell\ell'}$ or between different n shells if ℓ and ℓ' are equal, $\left\langle \psi_{n\ell} \psi_{n'\ell} \right\rangle = \delta_{nn'}$. Thus, ψ_{4f} is required to be orthogonal to ψ_{5f} and not to ψ_{5d} or ψ_{5g}. However, since the electron orbital angular momentum ℓ is not a good quantum number so close to the core electrons, an orthogonality between the approximating radial wave functions is probably more valid than if such a condition were not imposed.

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